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# Observation of multiply ionized plasmas with dominant bound electron contribution to the index of refraction

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## Abstract

We report anomalous fringe shifts observed in soft x-ray laser interferograms of laser-created Al plasmas. This clear experimental evidence shows that the contribution of bound electrons can dominate the index of refraction of laser-created plasmas at soft x-ray wavelengths, resulting in values greater than 1. The comparison of measured and simulated interferograms shows that this results from the dominant contribution of low charge ions to the index of refraction. This usually neglected bound electron contribution can affect the propagation of soft x-ray radiation in plasmas and the interferometric diagnostics of plasmas for many elements.

*Key words:* Interferometry; Soft x-ray laser; Index of refraction; Plasmas;  
Anomalous dispersion

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## 1 Introduction

In multiply ionized laser-created plasmas the index of refraction is usually calculated assuming that the contribution of bound electrons is negligible compared with that of free electrons. For example in all plasma interferometry experiments to date that used soft-x-ray laser probes, the spatial distribution of the electron density has been obtained assuming that the index of

refraction is determined by the density of free electrons[1, 2, 3, 4, 5, 6, 7]. In partially ionized plasmas two color interferometry is often used to separate the contribution to the index of refraction of neutral atoms and free electrons [8].

In this paper we present soft x-ray laser interferometry data obtained with a picosecond 14.7 nm (84.4 eV) laser probe that shows clear evidence of the dominant contribution of bound electrons to the index of refraction in the late stages of the evolution of an Al plasma created by a high power laser. The interferograms show that the fringes, late in the plasma evolution, in the periphery of the plasma and close to the target surface bend toward the target. In our experiment this direction of the fringe shifts is indicative of an index of refraction greater than 1 (hereafter referred as negative fringe shifts). In contrast, at earlier times all the fringes are observed to bend away from the target (positive fringe shifts). The observation of similar negative fringe shifts was previously reported from an independently realized Al laser-created plasma soft x-ray laser interferometry experiment at 13.9 nm [9]. Analysis of our data, with the assistance of hydrodynamic model simulations, indicates that late in the plasma evolution the contribution of bound electrons dominates the index of refraction, causing the observed negative fringe shifts in the periphery regions of the plasma. Herein we expand on results previously published in reference [10]. The significance of the result goes beyond the particular case of aluminum, as this effect can strongly affect the index of refraction of many ionized materials at soft x-ray laser wavelengths and needs to be carefully considered when analyzing experiments.

## 2 Experimental Setup

The experiment was performed using a transient 14.7 nm Ni-like Pd soft x-ray laser [11] combined with an amplitude division diffraction grating interferometer (DGI)[4, 7]. The soft x-ray laser was pumped by two beams from a chirped pulse amplification laser, the Compact Multipulse Terawatt (COMET) system at LLNL that operates at 1054 nm. An x-ray laser output of a few 10's of  $\mu\text{J}$  was achieved by optically pumping a polished Pd target with a sequence of a 600 ps long pulse ( $2\text{ J}$ ,  $2 \times 10^{11}\text{ Wcm}^{-2}$ ) and a 5 J energy short pulse of 6.7 ps or 13 ps (FWHM) duration at an incident intensity of  $6 \times 10^{13}\text{ Wcm}^{-2}$  and  $3 \times 10^{13}\text{ Wcm}^{-2}$  respectively. Traveling wave line focus excitation was achieved using a reflection echelon that consists of seven flat mirror segments placed before the focusing optics. Each mirror segment was offset by 0.12 cm to introduce the traveling wave toward the output of the laser with a delay of 7.7 ps per step. This results in a phase velocity of  $c$  along the line focus length and ensures that peak gain conditions are experienced by the propagating x-ray laser photons. The horizontal angular divergence of the soft x-ray laser

was measured to be 2.8 mrad. The near field and far field beam characteristics were studied and the laser parameters were optimized to obtain beam properties suitable to perform interferometry. The temporal properties of the x-ray laser have been recently measured with a fast x-ray streak camera under the same laser pumping conditions as the interferometry experiments [12]. For saturated x-ray laser output the x-ray duration is typically in the range of 4.5 - 5.2 ps. With the longer 13 ps pumping pulse, the measured x-ray duration is slightly longer at 5.9 ps [12]. Therefore, the interferograms obtained with this setup have picosecond resolution. This short pulse duration permits the acquisition of “snap-shots” of the rapidly evolving plasma, overcoming the blurring of interference fringes that occur when the electron density profile changes significantly during the duration of the probe pulse. The combination of the robust grating interferometer with the relatively high repetition rate picosecond soft x-ray laser (1 shot every 4 minutes) permitted the acquisition of sequences of interferograms that map the evolution of the electron density distribution in high density laser created plasmas.

The DGI is set in a skewed Mach-Zehnder configuration as shown in Figure 1. The light incident on the first grating (G1) is diffracted with approximately equal intensity in the zero and first orders. These two beams that form the two arms of the interferometer are reflected at 2.6 degrees grazing incidence angle toward a second  $7.6 \times 8.5 \text{ cm}^2$  diffraction grating (G2) using two 35 cm long Au-coated mirrors (L1 and L2). Grating G2 recombines the two beams such that they exit the interferometer propagating with a small angular difference, selected to produce fringes of the spacing required by the particular experiment. The gratings have a line spacing of 900 lines/mm with a blaze angle of 2 degrees. This grating diffracts the beam into a zero and a first order component of approximately the same intensity when operating at an incidence angle of 6 degrees. For the selected ruling and angle of incidence, the resulting angle between the zero and first diffracted orders is 5.2 degrees. This results in a distance between the two arms of the interferometer at the target position indicated in Fig.1 to be  $\sim 1 \text{ cm}$ . There, the phase object of interest, in our case a laser created plasma, is placed. The reflectivity of the gold-coated diffraction gratings is maximized by the small grazing incidence angle used, resulting in an efficiency of 25 % per order on each grating, giving a total throughput for the instrument, excluding the imaging optics, of  $\sim 12\%$ . More details on the DGI, including a discussion on the alignment procedure is discussed in reference [7].

The soft x-ray laser is imaged at the target position onto a  $1.33 \times 1.33 \text{ cm}^2$  charged-coupled device (CCD) with  $1024 \times 1024$  pixels of  $13 \times 13 \text{ }\mu\text{m}^2$  size [13]. The imaging optics consist of a 25 cm focal length Mo/Si multilayer spherical mirror (S1), set at 5 degrees off normal incidence, followed by an elongated Au-coated mirror (L3), positioned at a grazing angle of 9 degrees that relays the beam via a flat 45 degrees Mo/Si multilayer mirror to the CCD

detector located approximately 5.5 meters away from the imaging mirror. This imaging set up has a total magnification of  $22\times$ . The overall system spatial resolution was determined to be  $\sim 2\text{ }\mu\text{m}$ .

### 3 Results

Figure 2 shows interferograms of an expanding Al laser-produced plasma. The plasmas were heated at an irradiance of  $10^{13}\text{ Wcm}^{-2}$  by focusing a 600 ps, 3 J, 1054 nm laser into a 3.1 mm long  $\times$  12  $\mu\text{m}$  wide line focus on a 1 mm long flat Al target. The Al plasma was generated within the path of one arm of the interferometer, and the timing between the heating and probe beams was measured using fast photodiodes. The first four frames corresponding to the earlier time of the plasma evolution (0 to 0.9 ns) show a rapid lateral expansion of the plasma together with the formation of an on-axis density depression. At these times the fringes shift away from the target, even in the central region of the plasma where the on-axis depression is observed. A similar two-dimensional feature was observed in soft x-ray laser interferograms of laser-created Cu plasmas obtained using a 46.9 nm laser [14, 15]. In that case the simulations showed that the electron density minimum in the central region of the plasma is the result of pressure equilibrium between the irradiated plasma region and the low temperature side-lobes, created by plasma-radiation induced ablation of the surrounding target region. The last four frames in Fig. 2 show interferograms in which the region close to the target presents increased absorption, an indication of a colder plasma. Also the fringes at the periphery of the plasma shift toward the target, a phenomenon that requires a total index of refraction greater than 1.

In phase shift interferometry the number of fringe shifts, given by  $N_f = \frac{1}{\lambda} \int_0^L (1 - \eta) dl$ , is negative when the index of refraction ( $\eta$ ) of the plasma is greater than 1. The contribution of the free electrons to the plasma index of refraction is always less than 1, as determined by  $\eta = (1 - n_e/n_{crit})^{\frac{1}{2}}$  where  $n_{crit} = 5 \times 10^{24}\text{ cm}^{-3}$  is the critical density for  $\lambda = 14.7\text{ nm}$ . This suggests that the contribution of bound electrons to  $\eta$  is dominant and greater than one, in the region where negative fringe shifts are observed. The effect was not observed in interferograms of plasmas we generated with similar plasma heating beam conditions using several other target materials (Ti, Cr, Pd, Mo, Au). As an example, Fig. 3 shows interferograms for Al, Ti and Pd plasmas, obtained at approximately the same late time in the plasma evolution as the last frame in Fig. 2, 3 ns after the peak of the heating laser pulse. While the corresponding Al interferogram shows negative fringe shifts on the periphery of the plasma, the Ti and Pd interferograms show only positive fringe shifts. A possible explanation for this difference can be found reviewing the values of the neutral atom's real component of the scattering factor  $f_1^0$  at 14.7 nm,

as shown in Fig. 4. From all elements with atomic number ( $Z$ ) less than 54, Al is the only one that has a negative  $f_1^0$  [16, 17] due to the close proximity of the 84.4 eV photon energy to the Al L-shell absorption edge. Some elements with  $Z \geq 54$ , including I, Xe, Cs, Ba, Fr, Ra, Ac, also have a negative scattering factor at this wavelength. This negative scattering factor translates into an index of refraction greater than one ( $\eta = (1 - \frac{r_e \lambda^2 n_a f_1^0}{\pi})^{\frac{1}{2}}$  where  $n_a$  is the density of neutral atoms,  $\lambda$  is the probe beam wavelength and  $r_e$  is the classical electron radius). This suggests that significant densities of neutral Al atoms, for which the L-shell electrons have a significant contribution to  $\eta$ , will cause negative fringe shifts. However, the high photoionization cross section of neutral Al and the fact that the probe beam is not completely absorbed in the region where negative fringe shifts are observed indicate that the neutral density of this plasma should be negligible and that the negative fringe shifts are in fact caused by bound electrons on ions. Therefore, the real component of the index of refraction  $f_{1Z}^0$  was computed for the different ions present in the plasma. The calculation uses the best data available at this time with the possibility of further improvements in the future. We define the “effective”  $f_Z^*$  value as  $f_Z^{bound} + f_Z^{free}$  that includes the separate contributions of free and bound electrons, where  $f_Z^{free}$  is the number of free electrons corresponding to the ion of charge  $Z$  with the forward scattering supraindex (0) and real part (1) subindex omitted. We calculated the values for Al I - Al IX and are shown in Table 1. Details on this calculation can be found in [10, 18]. When the  $f_Z^*$  values differ from  $Z$  (the number of free electrons corresponding to that ion) the contribution of the bound electrons is significant. These  $f_Z^*$  values were used to calculate the total plasma index of refraction  $\eta = (1 - \frac{\sum_Z n_Z f_Z^*}{n_{crit}})^{\frac{1}{2}}$ , where  $n_Z$  is the density of ions with charge  $Z$  obtained from LASNEX simulations which self-consistently solve hydrodynamics and ionization kinetics equations [19]. All ions with charge up to  $Z=5$  were found to make a significant negative contribution to the index of refraction. The computed negative values of  $f_Z^*$  for Al I-Al IV suggest that the negative fringe shifts observed at the late times are due to the presence of a relatively high density of these low charge ions. Moreover, notice that ions up to Al VII make a significant contribution to the index of refraction. It should be noticed that this phenomenon does not require a close resonance and affects a broad range of wavelengths due to numerous bound-bound and bound-free transitions in many ionization stages. This is supported by the observation of negative fringe shifts in Al plasmas at two different probe beam wavelengths: 13.9 nm [9] and 14.7 nm (our experiments).

To confirm this interpretation of the data, the 2-dimensional (2-D) ion and electron density distributions for the plasmas of Fig. 2 was computed with LASNEX. The simulations were performed using flux-limited heat transport, multi-group radiation diffusion, and a detailed hydrogenic configuration atomic model with non-LTE physics for temperatures above 50 eV. The simulations

were conducted using the measured temporal and spatial profiles of the laser beam used to heat the plasma. Figure 5 shows 2-D maps of the computed electron density (contours) and the corresponding average ion charge distributions and Fig 6 shows the computed electron temperature 2-D maps. The electron density distributions measured during the early times of the expansion, including the lateral expansion and on-axis depression, are well reproduced by the code, and will be discussed in a future publication. This indicates that the assumption that the free electrons dominate the contribution to the index of refraction is valid at the early times, as is expected from the fact that at those times the plasma is hot and only highly ionized species are present. At later times, corresponding to the last two frames of Fig. 2, the simulations show that the degree of ionization in the plasma periphery decreases as the plasma cools, resulting in significant concentrations of Al II-Al IV ions present. The simulation results were used to synthesize the interferograms that are shown in Fig 7. These synthetic interferograms were calculated taking into account the contributions to the index of refraction from free electrons and Al ions using the computed “effective”  $f_Z^*$  factors and the electron density and ion density distributions computed with LASNEX. The synthesized interferogram corresponding to 0 to 0.9 ns delay show the central density depression and the lateral expansion observed in the measurements, and the late interferograms at 1.2 to 3 ns show the observed negative fringe shifts in the periphery of the plasma, in good agreement with the experiment. In the simulations, the negative fringe shifts appear slightly earlier (at 0.8 ns) than in the experiment (1.1 ns).

It should be expected that interferograms of the late stages of the evolution of many other laser-created plasmas involving other materials will also be significantly affected by the contribution of bound electrons. Moreover, the importance of the bound electron contribution is not always limited to plasmas with a low mean ion charge. For example, ten times ionized Pd atoms are computed to have an  $f_1^*$  value that is several times that of the corresponding number of free electrons, with the precise value of  $f_1^*$  depending strongly on the position of the resonance line[18]. It is also important to realize that the relative contribution of bound electron scattering is not always clearly evident in soft x-ray interferograms because at these wavelengths the free and bound electrons often contribute with the same sign to the phase delay. While soft x-ray laser interferometry allows measurements of higher plasma densities because of better spatial resolution, reduced absorption and reduced deflection angles within steep plasma density gradients, neglecting the contribution of bound electrons to the index of refraction could, in some cases, constitute a significant systematic error in the determination of the electron density.



## 4 Conclusions

In conclusion, we have demonstrated that bound electrons can contribute significantly to the index of refraction of multiply ionized plasmas at soft x-ray wavelengths in the vicinity of absorption edges. In the case of an Al plasma the effect causes a dramatic change of sign of the fringe shifts in interferograms acquired using  $\lambda = 14.7$  nm radiation. The significance of the result goes beyond the particular case of aluminum, as it can significantly affect the index of refraction of many materials at soft x-ray laser wavelengths.

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Fig. 1. Experimental setup, depicting the Diffraction Grating Interferometer. The soft x-ray laser enters and exits the interferometer through the lower right side.

Fig. 2. Sequence of soft x-ray laser interferograms ( $\lambda = 14.7$  nm) of Al line focus plasmas. The plasmas were generated by a 3 J heating beam focused into a  $12 \mu\text{m} \times 3.1$  mm line focus. The targets were 1 mm long. The times are measured respect to the peak of the 600 ps heating pulse. Fringes that bend toward the target (negative fringe shifts) are observed in the last two frames in the periphery of the plasma and close to the target.

Fig. 3. Comparison of interferograms obtained 3 ns after the peak of the heating laser pulse for Al, Ti and Pd targets for heating conditions similar as those used to obtain the Al interferograms in Fig. 2.

Fig. 4. Scattering factor for neutral atoms  $f_l^\theta$  vs Element's Atomic Number at 84.4 eV taken from Henke et al. [16] and Gullikson et al. [17]

Fig. 5. Sequence of simulated electron density contours and mean ionization distribution maps computed using the LASNEX code for an Al plasma created with the irradiation conditions of Fig. 2.

Fig. 6. Sequence of simulated electron density contours and Electron temperature distribution maps computed using the LASNEX code for an Al plasma created with the irradiation conditions of Fig. 2.

Fig. 7. Synthesized interferograms computed using the calculated electron and ion densities from Fig. 5 and the calculated scattering factors for Al ions.

Table 1

Contribution of the free and bound electrons to the index of refraction.

Ion species	Effective Scattering factor $f_Z^*$
Al I	-0.85
Al II	-4.19
Al III	-3.54
Al IV	-1.80
Al V	0.84
Al VI	3.54
Al VII	5.30
Al VIII	6.73
Al IX	8
Al X	9.18
Al XI-Al XIV	# of free electrons

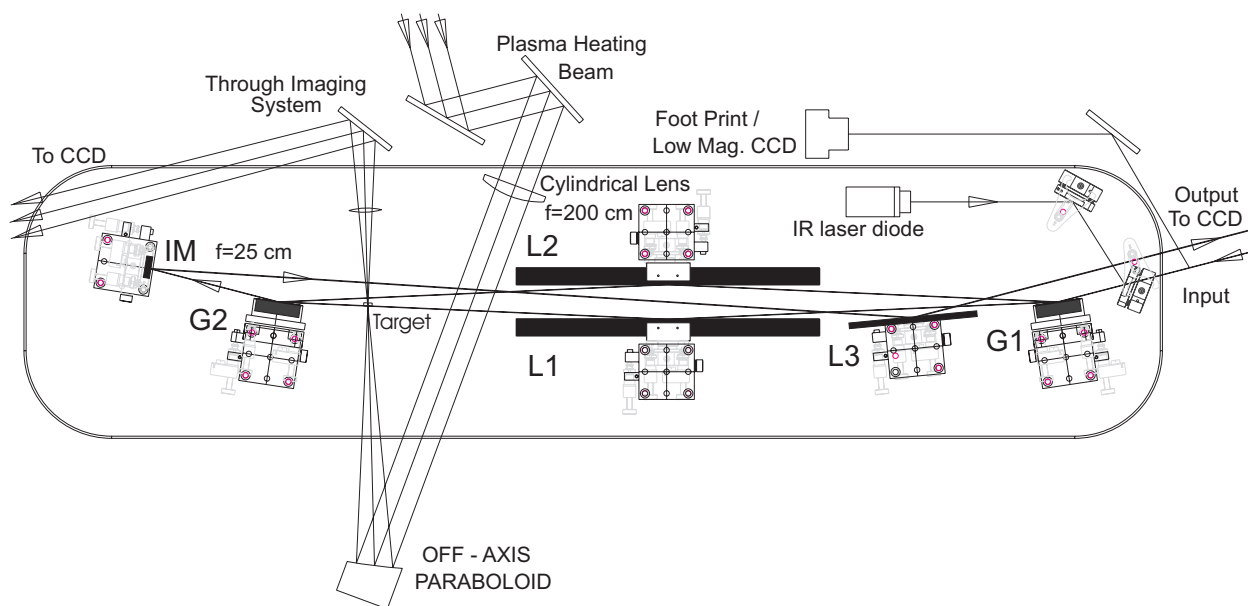
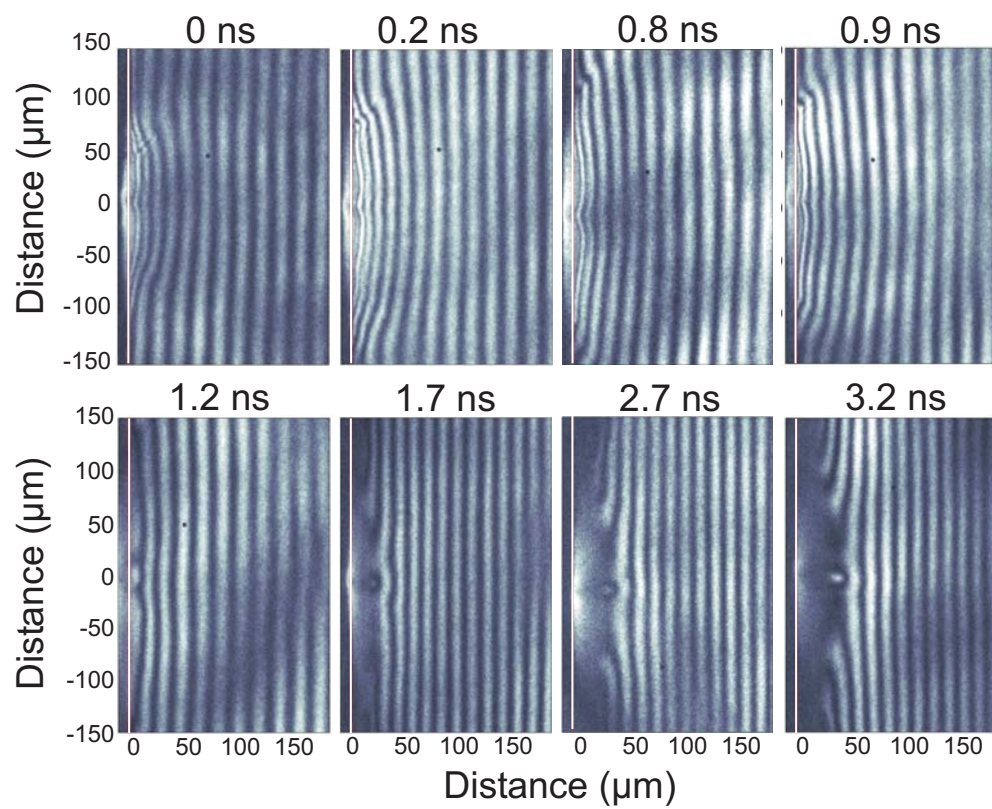


Figure 1



Aluminum

Titanium

Palladium

